1 Introduction

It is of importance to first define deflagrations and detonations and give the characteristics that distinguish these two types of combustion waves. Since this book is concerned with a description of the detonation phenomenon, it is of value to first introduce the various topics that are concerned with detonations prior to their detailed description in later chapters. In this manner, a global perspective can be obtained and permit selective reading of the chapters for those who are already familiar with the subject.

In telling a story, it is natural to start from the beginning, and thus the presentation of the various topics follows more or less their historical development. However, no attempt is made here to discuss the extensive early literature. A historical chronology of detonation research covering the period from its first discovery in the late 1800s to the state of knowledge in the mid 1950s has been documented by Manson and co-workers (Bauer *et al.*, 1991; Manson & Dabora, 1993). An extensive bibliography of the early works is given in these two papers for those who want to pursue further the history of detonations. This chapter is in essence a qualitative summary of the material covered in this book.

1.1. DEFLAGRATIONS AND DETONATIONS

Upon ignition, a combustion wave propagates away from the ignition source. Combustion waves transform reactants into products, releasing the potential energy stored in the chemical bonds of the reactant molecules, which is then converted into internal (thermal) and kinetic energy of the combustion products. Large changes in the thermodynamic and gasdynamic states occur across the combustion wave as a result of the energy released. The gradient fields across the wave generate physical and chemical processes that result in the self-sustained propagation of the combustion wave.

Generally speaking, there are two types of self-propagating combustion waves: deflagrations and detonations. *Deflagration* waves propagate at relatively low subsonic velocities with respect to the reactants ahead of it. As subsonic waves, disturbances downstream can propagate upstream and influence the initial state of the reactants. Thus, the propagation speed of a deflagration wave depends not only on the properties and initial state of the explosive mixture, but also on the rear boundary condition behind the wave (e.g., from a closed-end or an open-end tube). A deflagration is an expansion wave where the pressure drops across the reaction front, and the combustion products are accelerated away from the wave in a direction opposite to its propagation. Depending on the rear boundary condition (e.g., for a closed-end tube the particle velocity is zero in the products), the expansion of the products causes a displacement of the reactants ahead of the reaction front. Thus, the reaction front propagates into reactants that are moving in the direction of propagation. The deflagration speed (with respect to the fixed laboratory coordinates) will then be the sum of the displacement flow velocity of the reactants and the velocity of the reaction front relative to the reactants (i.e., the burning velocity). Compression waves (or shocks) are also formed in front of the reaction front as a result of the displacement flow. Thus, a propagating deflagration wave usually consists of a precursor shock followed by the reaction front. The strength of the precursor shock depends on the displacement flow velocity, and hence on the rear boundary condition.

The mechanism by which the deflagration wave propagates into the reactants ahead of it is via diffusion of heat and mass. The steep temperature and chemical species concentration gradients across the reaction front result in the transport of heat and radical species from the reaction zone to the reactants ahead to effect ignition. Therefore, a deflagration is essentially a diffusion wave, and as such, it has a velocity proportional to the square root of the diffusivity and of the reaction rate (which governs the gradients). If the deflagration front were turbulent, we may, within a one-dimensional context, define a turbulent diffusivity to describe the transport processes. A *flame* is generally defined as a stationary deflagration wave (with respect to laboratory coordinates) stabilized on a burner with the reactants flowing toward it. However, the term *flame* is often also used for the reaction front even in a propagating deflagration wave.

A *detonation* wave is a supersonic combustion wave across which the thermodynamic states (e.g., pressure and temperature) increase sharply. It can be considered as a reacting shock wave where reactants transform into products, accompanied by an energy release across it. Because the wave is supersonic, the reactants ahead are not disturbed prior to the arrival of the detonation; hence they remain at their initial state. Because it is a compression shock wave, the density increases across the detonation, and the particle velocity of the products is in the same direction as that of the wave motion. The conservation of mass then requires either a piston or expansion waves to follow the detonation front. For a piston-supported detonation (known as a strong or overdriven detonation), the flow can be subsonic behind the detonation, since no expansion waves trail behind it. However, for a freely propagating detonation (without a supporting piston motion behind it), the expansion waves behind the detonation front will reduce the pressure and particle velocity to match the rear boundary condition. Since the flow is subsonic behind a strong detonation, any expansion wave will penetrate the reaction zone and attenuate the detonation. Thus, a freely propagating detonation must have either a sonic or a supersonic condition behind it. Detonations with a sonic condition behind them are called *Chapman–Jouguet* (CJ) detonations; those with a supersonic condition are called *weak* detonations. Weak detonations require special properties of the Hugoniot curve (i.e., curve representing the locus of equilibrium states of the detonation products for different detonation velocities) and are not commonly realized. Therefore, freely propagating detonations are generally CJ detonations with a sonic condition behind them.

Ignition of the reactants is effected by the adiabatic compression of the leading shock front that precedes the reaction zone of the detonation wave. An induction zone usually follows the leading shock where dissociation of the reactants and the generation of free radical species occur. The variation in the thermodynamic state in the induction zone is usually small. Following the induction zone, rapid recombination reactions occur with an accompanying temperature increase from the exothermic reactions. The pressure and the density drop through the reaction zone. Thus, the reaction zone of a detonation is similar to a deflagration wave, and a detonation wave is often considered to be a closely coupled shock–deflagration complex, except that ignition is due to adiabatic heating by the leading shock. The rapid pressure drop in the reaction zone, together with a further pressure decrease in the expansion waves that follow a freely propagating detonation, provides the forward thrust that supports the leading shock front. Thus, the classical mechanism of propagation of an unsupported detonation is autoignition by the leading shock front, which in turn is driven by the thrust from the expanding products in the rear.

Self-propagating deflagrations are intrinsically unstable, and there exist numerous instability mechanisms that render the reaction front turbulent, thereby increasing its propagation speed. Thus, self-propagating deflagrations accelerate, and when boundary conditions permit, they undergo an abrupt transition to detonations. Prior to transition to detonation, turbulent deflagrations can reach high supersonic speeds (relative to a fixed coordinate system). By *high-speed* deflagrations we usually mean these accelerating deflagrations during the transition period. When detonations propagate in very rough-walled tubes, their propagation speeds can be substantially less than the normal CJ velocity. These low-velocity detonations are referred to as “*quasi-detonations*.” The velocity spectra of high-speed deflagrations and quasi-detonations overlap. The complex turbulent structure of these waves is similar, suggesting that their propagation mechanisms may also be similar. Thus, it is difficult to draw a sharp distinction between them.

The different types of combustion waves described in this section manifest themselves under different initial and boundary conditions. Their consideration is the subject of this book.

Figure 1.1. Marcelin Berthelot (1827–1907) and Paul Vieille (1854–1934).

1.2. DISCOVERY OF THE DETONATION PHENOMENON

It has been known since the fifteenth century that certain chemical compounds (e.g., mercury fulminate) undergo unusually violent chemical decomposition when subjected to mechanical impact or shock. However, it was not until the development of diagnostic tools, which permitted the rapid combustion phenomenon to be observed and the propagation velocity of the combustion wave to be measured, that we can say the detonation phenomenon was discovered. Abel (1869) was perhaps the first to measure the detonation velocity of explosive charges of guncotton. However, it was Berthelot and Vieille (Berthelot, 1881; Berthelot & Vieille, 1883) who systematically measured the detonation velocity in a variety of gaseous fuels (e.g., H_2, C_2H_4, C_2H_2) mixed with various oxidizers (e.g., O_2, NO, N_2O_4) and diluted with various amounts of inert nitrogen, thereby confirming the existence of detonations in gaseous explosive mixtures.

Mallard and Le Châtelier (1883) used a drum camera to observe the transition from deflagration to detonation, thus demonstrating the possibility of two modes of combustion in the same gaseous mixture. They also suggested that the chemical reactions in a detonation wave are initiated by the adiabatic compression of the detonation front. Therefore, in the late 1800s, supersonic detonation waves in gaseous explosive mixtures were conclusively demonstrated to be distinctly different from slowly propagating deflagration waves. The early pioneers (Berthelot and Vieille; Dixon, 1893, 1903) all recognized the role played by adiabatic shock compression in initiating the chemical reactions in a detonation wave.

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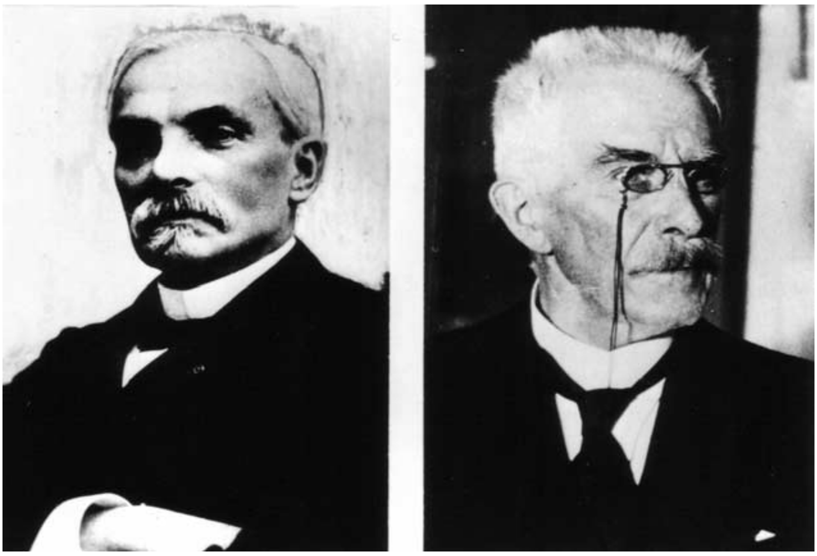

Figure 1.2. Ernest Mallard (1833–1899) and Henry Le Châtelier (1850–1936).

1.3. CHAPMAN–JOUGUET THEORY

A quantitative theory that predicts the detonation velocity of an explosive mixture was formulated by Chapman (1889) and Jouguet (1904, 1905) shortly after the discovery of the phenomenon.

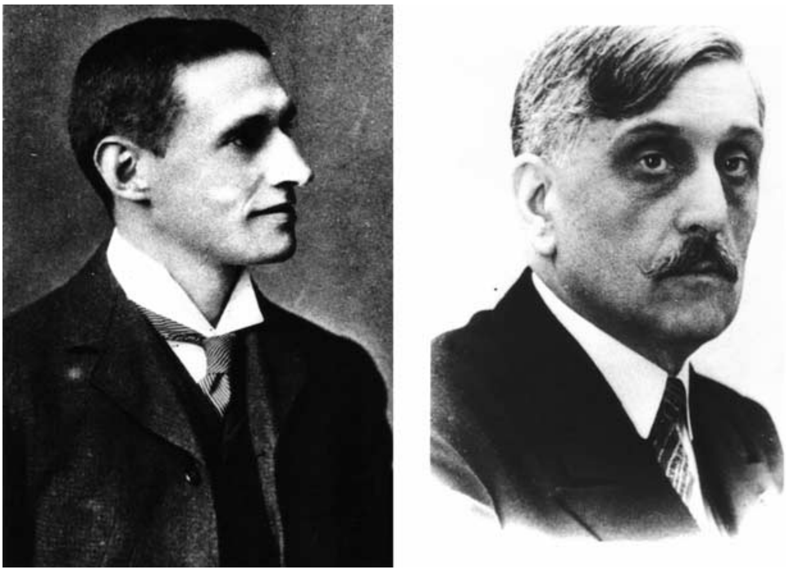

Figure 1.3. Donald Leonard Chapman (1869–1958) and Ehrile Jouguet (1871–1943).

Both Chapman and Jouguet based their theory on the works of Rankine (1870) and Hugoniot (1887, 1889), who analyzed the conservation equations across a shock wave. For a detonation wave, the transformation of the reactants into products across the wave results in the release of chemical energy. Assuming equilibrium downstream of the wave, it is possible to determine the chemical composition of the products in terms of the thermodynamic state, and thus the chemical energy released across the detonation can be determined. Unlike a non-reacting shock wave, two possible solutions exist for a given detonation wave speed: the strong and weak detonation solutions. The pressure and density of the strong detonation solution are greater than those of the weak detonation solution. The flow downstream of a strong detonation is subsonic (relative to the wave), whereas for a weak detonation, it is supersonic. The two solutions converge when the detonation velocity is a minimum. No solution exists for detonation velocities below this minimum value. Since a continuous spectrum of detonation velocities above the minimum is possible for a given explosive mixture, the task of a detonation theory is to provide a criterion for the choice of the appropriate detonation velocity for an explosive mixture at given initial conditions.

Chapman's criterion is essentially to choose the minimum-velocity solution. The argument he provided was simply that for a given explosive mixture, experiments indicate that a unique detonation velocity is observed. Thus, the minimum-velocity solution must be the correct one. Jouguet, on the other hand, investigated the locus of the thermodynamic states for various detonation velocities (i.e., the Hugoniot curve). He determined the entropy variation along the Hugoniot curve and discovered a minimum. He further noted that the minimum entropy solution corresponds to sonic condition downstream of the detonation. Jouguet then postulated that the minimum-entropy solution (the sonic solution) is the appropriate one to choose. His collaborator, Crussard (1907), later showed that the minimum-velocity solution corresponds to the minimum-entropy solution and also gives sonic flow downstream of the wave. Thus, both Chapman and Jouguet provided a criterion (i.e., minimum velocity or minimum entropy) for the choice of the appropriate detonation velocity for a given explosive mixture, and this is now referred to as the CJ theory. Neither Chapman nor Jouguet provided physical or mathematical justification for their postulates.

It is of interest to note that Mikelson (1890) in Russia had earlier developed a similar theory for detonation. He also analyzed the conservation equations across a detonation and found the existence of two possible steady solutions that converge to a single solution when the detonation velocity is a minimum. Unfortunately, his doctoral dissertation, where his analyses were reported, was not known outside Russia. Although these three researchers had independently formulated a gasdynamic theory of detonation at about the same time, only Chapman's and Jouguet's names are associated with the theory.

Note that the CJ theory is incomplete until more rigorous physical or mathematical arguments are provided to justify the criterion for the selection of the solution.

A few subsequent researchers had based their arguments on entropy (Becker, 1917; 1922a, 1922b; Scorah, 1935). But Zeldovich (1940/1950) had refuted any thermodynamic argument by pointing out that the entropy increase across a shock wave alone does not imply that the shock wave will exist. The mechanism for generating the shock wave (e.g., piston motion behind it) is required. The justification for the CJ criterion used by the early investigators was based on the properties of the solution of the Rankine–Hugoniot equations across the detonation front (e.g., minimum velocity, minimum entropy, or sonic conditions). G.I. Taylor (1950), who studied the dynamics of the detonation products behind the front, was the first to point out that the boundary condition at the front must lead to a physically acceptable solution for the non-steady expansion flow of the combustion products behind the detonation. For planar detonations, the Riemann solution is compatible to the sonic conditions of a CJ detonation. However, for spherical detonations, a singularity in the form of an infinite expansion gradient is obtained when the CJ conditon is imposed. This has led to a controversy over the existence of steady CJ spherical detonations (e.g., Courant and Friedrichs, 1948; Jouguet, 1917; Zeldovich and Kompaneets, 1960). The strong detonation solution can be eliminated for freely propagating detonations, because the conservation of mass requires that an expansion wave must follow the detonation to reduce the density. Because the flow is subsonic behind the strong detonation, the expansion waves will penetrate the reaction zone and attenuate the detonation. However, the weak detonation solution is more difficult to eliminate. It was von Neumann (1942) who provided an interesting argument for rejecting the weak detonation solution by examining the structure of the detonation wave. He first assumed that intermediate Hugoniot curves can be constructed based on a given degree of completion of the chemical reactions. He then showed that if the intermediate Hugoniot curves do not intersect one another, then the weak detonation solution cannot be attained. However, if the chemical reactions are such that the intermediate Hugoniot curves do intersect, he showed that weak detonations are possible. Such detonations are referred to as *pathological* detonations and do exist for certain explosives with a temperature overshoot. It may be concluded that a gasdynamic theory based only on the Rankine–Hugoniot reactions across the front cannot justify the CJ criterion. Both the solution for the nonsteady flow of the detonation products and the nature of the chemical reactions within the structure must be considered in the selection of the appropriate solution of the Rankine–Hugoniot equations.

1.4. THE DETONATION STRUCTURE

The CJ theory completely bypasses the details of the detonation structure (i.e., the transition processes from reactants to products). It is essentially a consideration of the possible solutions of the steady one-dimensional conservation equations that link the upstream and downstream equilibrium states of the reactants and products, respectively. Without a description of the structure, the propagation mechanism of

Figure 1.4. Yakov B. Zeldovich (1914–1987), John von Neumann (1903–1957), and Werner Döring (1911–2006).

the detonation wave cannot be known. Although ignition via shock compression was known to the early pioneers who discovered the phenomenon, it was Zeldovich (1940), von Neumann (1942), and Döring (1943) who explicitly described the model of the detonation structure as comprising a leading shock front followed by a chemical reaction zone.

Due to the Second World War during the early 1940s, we may assume that these three researchers were unaware of each other's work. In his original paper, Zeldovich included heat and momentum losses within the structure to investigate their effects on the propagation of the detonation wave. An important consequence of the loss terms is that the integral curve encounters the sonic singularity prior to chemical equilibrium. Seeking a regular solution across the sonic singularity requires a unique value for the detonation velocity, and thus, the term *eigenvalue detonation* is often used in modern literature. With heat and momentum losses, the detonation velocity is less than the equilibrium CJ value. At some critical values for the loss terms, no steady solution can be obtained, which can be interpreted as the onset of the detonation limits observed experimentally. Heat and momentum losses to the walls are two-dimensional effects, and to model them as one-dimensional gives an incorrect description of their physical effects on the detonation structure. Nevertheless, Zeldovich's analysis led to an important mathematical criterion for determining the detonation solution, namely, regularity at the sonic singularity.

Von Neumann's analysis of the detailed transition processes in the detonation structure is an attempt to provide a more rigorous justification for the Chapman–Jouguet criterion, in particular the elimination of the weak detonation solution. He introduced a parameter n to denote the progress of the chemical reaction from the leading shock to the final products, with $0 \leq n \leq 1$. At each value of n, he assumed equilibrium states $(p(n), v(n))$ can be defined, permitting an intermediate Hugoniot curve (i.e., the locus of states that satisfy the conservation equations for a fixed value of n) to be constructed. Then, from the geometry of these intermediate Hugoniot curves, he demonstrated that weak detonations are not possible in general if the intermediate curves do not intersect one another. However, for certain reactions, where the Hugoniot curves do intersect, the detonation velocity obtained is higher than the equilibrium CJ value, and the solution itself lies on the weak detonation branch of the equilibrium Hugoniot curve, where $n = 1$. The importance of von Neumann's analysis is the demonstration of pathological detonations, which have velocities higher than the equilibrium CJ value. These pathological detonations are observed experimentally when there exists a temperature overshoot in the chemical reaction process toward equilibrium.

Werner Döring had studied under Richard Becker, who carried out important fundamental work on shock and detonation waves throughout the 1920s and 1930s. Becker had already conceived the idea that the detonation structure is in essence a shock wave where chemical transformation takes place. For this reason, Becker thought that heat conduction and viscosity effects could be important. As it turns out, chemical reactions occur much later downstream, and the leading shock can then be dissociated from the reaction zone.

Döring's analysis of the detonation structure is remarkably similar to that of von Neumann. He defined a reaction progress variable n (in terms of the concentrations of the reactants), which goes from 0 to 1 as the reaction proceeds toward equilibrium. He integrated the conservation equations across the reaction zone and obtained the profiles for the thermodynamic states within the detonation zone. In honor of the three researchers who carried out the analysis of the structure of the detonation, the model of a shock followed by chemical reactions is now referred to as the Zeldovich–von Neumann–Döring (ZND) model. The ZND model now provides the mechanism responsible for the propagation of the detonation wave, namely, ignition by adiabatic compression across the leading shock, which is in turn maintained by the thrust generated by the expansion of the gases in the reaction zone and in the products.

It should be noted that the CJ criterion that selects the minimum velocity solution is only a postulate and does not follow from the conservation laws across the detonation front. The minimum velocity solution implies that the Rayleigh line is tangent to the equilibrium Hugoniot curve, and therefore, the sonic condition is based on the equilibrium sound speed. In the alternate method where the ZND equations are integrated across the structure of the front, the criterion used in iterating for the desired detonation velocity is the regularity condition at the sonic singularity. The sonic condition is now based on the frozen sound speed. Although the solution still lies on the equilibrium Hugoniot curve, it is no longer the minimum-velocity (or tangency) solution and now lies on the weak branch of the equilibrium Hugoniot curve. There is no reason to expect that the two solutions are the same since the method and the criterion used to obtain them are different. The use of the CJ criterion is simpler since the details of the reaction zone are not involved. The detonation velocity can be found from computations using the equilibrium thermodynamic properties of the reacting mixture. On the other hand, integration across the ZND structure is rather involved and requires a knowledge of the detailed chemical kinetics of the reactions. However, solutions for *pathological detonations* can now be obtained. It is difficult to determine which solution corresponds to reality since the two detonation velocities differ slightly by only a few percent. Furthermore, the nonsteady three-dimensional cellular structure of real detonations and the influence of the boundary conditions on the propagation of the detonation wave probably have larger effects on the detonation velocity. In view of the relative ease in carrying out an equilibrium thermodynamic calculation, the CJ criterion that selects the minimum velocity solution based on tangency of the Rayleigh line to the equilibrium Hugoniot curve is generally used to find the detonation velocity of a given explosive mixture.

1.5. DYNAMICS OF THE DETONATION PRODUCTS

The analysis of the nonsteady flow of the detonation products is as important as the study of the conservation equations across the detonation front. Solutions for the flow behind planar and spherical detonations were first obtained by G.I. Taylor (1940/1950) and also independently by Zeldovich (1942). Taylor pointed out the important fact that a steady detonation is only possible if a solution for the nonsteady flow in the products can be found that can satisfy a steady state boundary condition at the CJ detonation front. For the planar case, the Riemann solution can be matched to the condition behind a CJ detonation. Thus, steady planar CJ detonations are possible. However, for diverging cylindrical and spherical detonations, it is found that there would exist a singularity in the form of an infinite expansion gradient behind the detonation if the sonic condition of a CJ wave were to be imposed. Such a singularity does not exist for strong or weak detonations. However, strong and weak detonations can be ruled out for other reasons. The infinite expansion singularity obtained behind the front raises a question as to the existence of steady cylindrical and spherical detonations. It is clear that if we were to consider the reaction zone thickness to be finite, then steady diverging CJ detonations cannot exist. This is due to the influence of curvature on the flow in the reaction zone, which leads to a detonation velocity less than the equilibrium CJ velocity. Since curvature varies with radius, the detonation velocity will change as it expands and will only reach the CJ value asymptotically at infinite radius. Lee *et al.* (1964) also pointed out that the direct initiation of spherical detonations requires a substantial amount of energy by the ignition source (Laffitte, 1923; Manson and Ferrie, 1952; Zeldovich *et al.*, 1957). If the initiation energy is considered, then a strong blast wave is generated at small radius and a CJ detonation would only be obtained asymptotically at infinite radius. Thus, both the consideration of a finite reaction zone thickness and the inclusion of the initiation energy led to the conclusion that steady CJ spherical detonations are not possible. Furthermore, the instability of the detonation front leads to a transient three-dimensional cellular structure, which differs from the one-dimensional structure assumed in the analysis of G.I. Taylor and Zeldovich. Therefore, the gas-dynamic theory of detonation based on a consideration of just the conservation laws across the front (i.e., Rankine–Hugoniot equations) is incomplete. Both the